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Removal of Radiostrontium by Leaching, Runoff, and Plant Uptake as Influenced by Soil and Crop Management Practices

F. HAGHIRI and F. L. HIMES

OHIO AGRICULTURAL RESEARCH AND DEVELOPMENT CENTER
WOOSTER, OHIO

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Removal of Radiostrontium by Leaching, Runoff, and Plant Uptake as Influenced by Soil and Crop Management Practices[1]

F. HAGHIRI and F. L. HIMES[2]

INTRODUCTION

The biological importance of strontium-90 in the food chain has been known for more than 2 decades. The fate of soil-applied strontium-90 is determined by a number of processes, such as soil fixation and removal by plant uptake, leaching, and runoff. In order to predict the magnitude of loss, accumulation, or activity of strontium-90 in a given site, quantitative evaluation of the effects of the above processes under natural environmental conditions is essential.

Field investigations were conducted to determine the effects of management practices on the removal of strontium-90 from soil by leaching, runoff, and plant uptake. This bulletin is a report of the findings over a 10-year period (1962-1972).

FIELD RESEARCH SITE

The field research area occupies a small isolated watershed, about 1.5 acres, near the Ohio Agricultural Research and Development Center, Wooster.

Thirty field microplots (0.002 acre each) were designed and constructed to provide isolation of the soil of each plot and of the vegetation grown (Fig. 1). Facilities were provided for isolation, collection, and measurement of runoff and leachate water from each plot (2).

The plots are located on a moderately eroded Canfield silt loam soil. Canfield silt loam, in undisturbed condition, is strongly to very strongly acid in the upper part of the profile. The research plots are located in a cultivated area and the soil is less acid than the undisturbed soil due to liming (Table 1). The predominant clay mineral of the Canfield silt loam soil is illite.

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[1]This research was performed under Contract No. AT (11-1)-414 with the U. S. Atomic Energy Commission.

[2]The authors are professors in the Department of Agronomy, Ohio Agricultural Research and Development Center and The Ohio State University.

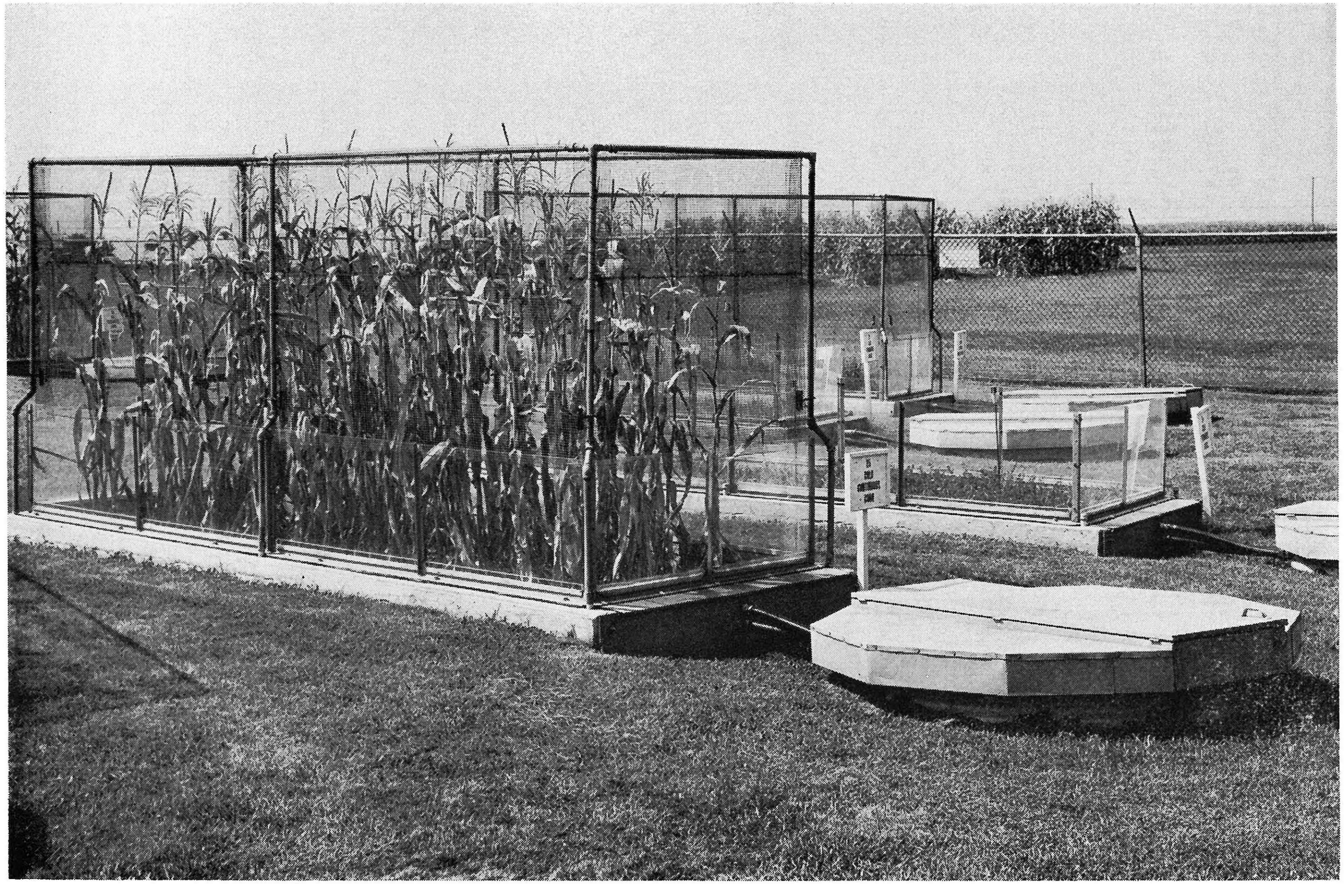

FIG. 1.—A general view of the experimental site showing microplots and catchment basins.

One important property of this soil is the presence of a fragipan (B_x horizon) in the lower subsoil. This relatively dense horizon appreciably reduces permeability to air and water and restricts the deeper penetration and distribution of root systems of growing plants.

EXPERIMENTAL PROCEDURES

In June 1962, all microplots were adequately fertilized with nitrogen, phosphorus, and potassium according to soil test. Five treatments were used: 1) crop rotation (corn, wheat, 3 years of meadow) with low rates of lime; 2) same crop sequence with high rates of lime; 3) permanent grass mixture (orchardgrass, bromegrass, and timothy); 4) gravel mulch with no vegetation; and 5) continuous corn. Treatments were replicated three times.

Low lime and *high lime* rotation plots were limed with 1,000 and 8,000 lb. of $CaCO_3$ per acre, respectively. Prior to planting, fertilizers and lime were broadcast and incorporated into the top 6 inches of soil (Table 2). After the first year, all plots with the exception of gravel

TABLE 1.—Organic Matter Content, Cation Exchange Capacity, Exchangeable Bases, Base Saturation, and pH Range of Soils of Plot Area.*

Horizon	Depth (Inches)	Percent OM	CEC meq/100 g.	Exchangeable Bases meq/100 g. Ca	Mg	K	Percent Base Saturation	pH Range
Ap	0-6	1.8	12.9	5.9	1.3	0.22	58	5.4-7.0
B_1	6-8	0.8	9.8	3.9	1.1	0.16	52	5.0-6.8
B_2	8-16	0.5	14.3	4.1	1.8	0.19	43	4.6-5.7
B_x Upper	16-45		16.4	4.9	2.7	0.18	43	4.6-4.8
B_x Lower	16-45		13.9	6.3	3.4	0.14	70	6.0-6.3
B_3	45-56		12.8	6.0	2.6	0.12	72	6.4-6.9

*Data obtained by N. Holowaychuk, Department of Agronomy, OARDC and OSU. Averaged values of five profiles.

TABLE 2.—pH and Exchangeable Calcium of Canfield Soil (0 to 4-Inch Depth) After Fertilizer and Lime Application (1962).

Cropping System	pH (Soil/Water = 1:2.5)	Calcium (meq/100 g. soil)
Rotation low lime	5.3	7.9
Rotation high lime	7.1	17.0
Gravel mulch	5.0	6.9
Permanent grass	6.1	10.6
Continuous corn	6.5	13.2

mulch were fertilized annually. Grass mixtures were top-dressed with N, P, and K; alfalfa was top-dressed with P and K; and corn was fertilized by band placement of N, P, and K.

Shortly after planting on June 21, 1962, 15 microplots were treated with carrier-free strontium-90 ($SrCl_2$ solution) at a rate of 21.64 μci per plot to simulate radioactive fallout. The remaining plots have been kept untreated to serve as control. Soon after the application of strontium-90, six bare plots (three control and three treated plots) were mulched with 2 inches of acid-washed gravel (approximately ¼ inch in diameter).

Soil sampling frequency from 1962-1967 was once a year and from 1967-1971 was once every 2 years. From 1962 to 1971, and from 1964 to 1971, composite soil samples were obtained from the 0 to 4 and 4 to 8-inch depths, respectively, to determine the possible movement of strontium downward with time. In addition, in 1969 and 1971, soil samples from the 8 to 12-inch depth were obtained.

Total amounts of runoff (water and sediment) and leachate water from each microplot were determined annually. Runoff was channeled from the microplot through a flume pipe to a calibrated catchment basin (300 gal. capacity), located at the lower end of each plot. Runoff volume was determined after each rainfall. A composite sample of runoff water and sediment was obtained from each plot by an electrically operated pendulum sampler located in each catchment basin. The sampler was designed to collect approximately 2% of the total runoff in a container inside the catchment basin.

To collect leachate from the plots, a perforated copper tube drain outlet was installed at the lower end of each plot just above the fragipan horizon. The drain outlet from each plot was extended into a leachate cellar where the leachate from each plot was collected in a corresponding 27-gallon calibrated tank. Each leachate sample was composed of an annual composite of subsamples (100-ml. aliquots) of leachate water taken each time water percolated through the soil profile. Runoff (water plus sediment) and leachate water samples were analyzed for strontium-90 activity.

All cut plant materials were removed from the microplots. Permanent grass plots were harvested on 42-day cutting schedules. Alfalfa crops were harvested at bloom stage. The number of cuttings for both grass and alfalfa varied from 1 year to another, depending on the amount of precipitation and growth. Other crops, such as corn and wheat, were harvested at maturity. Alfalfa, grass, corn fodder, shelled corn, wheat straw, wheat hulls, and wheat grain samples were analyzed for strontium-90 and calcium contents.

The strontium-90 content values of various samples obtained from strontium-90 treated plots were corrected for the decay of strontium-90. The strontium-90 concentration values of different materials from the control plots were used to correct strontium-90 from natural fallout intercepted by the treated plots.

RESULTS AND DISCUSSION

The net rate of strontium-90 removal by various processes from Canfield silt loam soil (0 to 4-inch depth) under different soil and crop management practices decreased with time. The relationship between log strontium-90 and log time at the 0 to 4-inch depth was linear under all cropping systems. At this depth, the net rate of strontium-90 loss was maximum under gravel mulch and minimum under permanent grass cropping systems (Fig. 2). The calculated linear regression equations of log strontium-90 on log time and R^2 values for rotation *low lime,* rotation *high lime,* and continuous corn cropping systems were: $Y = 4.48 - 0.45X$, $R^2 = 0.80^{**}$; $Y = 4.46 - 0.34X$, $R^2 = 0.76^{**}$; and $Y = 4.47 - 0.45X$, $R^2 = 0.74^{**}$. respectively. Although the strontium-90 content of the soil samples obtained from the 4 to 8-inch depth increased with time during 1964-1971, there were no significant differences in the rate of strontium-90 movement from the 0 to 4-inch depth to the 4 to 8-inch depth among the treatments.

Table 3 shows the percent distribution of strontium-90 (1971) in the top 12 inches of soil profile under various treatments. Ten years after the application of strontium-90, the concentration of this isotope at the 8 to 12-inch depth under gravel mulch was more than 12 times higher than the concentration under the permanent grass system. This was due to the larger volume of water which had percolated through the soil profile, and lower exchangeable calcium (Table 2) under gravel mulch conditions.

TABLE 3.—Distribution of Strontium-90 in the 0 to 12-Inch Soil Depth as Influenced by Liming and Cropping Systems (1971).

Cropping System	Percent ^{90}Sr‡		
	0-4 in.	4-8 in.	8-12 in.
Gravel mulch	61.0	30.0	9.0
Continuous corn	71.4	24.0	4.6
Rotation low lime*	71.2	23.1	5.7
Rotation high lime†	79.6	18.0	2.4
Permanent grass	82.0	17.3	0.7

*1000 lb. $CaCO_3$/acre applied in 1962 and 1966
†8000 lb. $CaCO_3$/acre applied in 1962 and 1966.
‡Average of three replications.

The effect of lime on the movement of strontium-90 in the soil profile can also be seen by comparing the strontium-90 concentrations of soil samples at the 8 to 12-inch depth from rotation *low lime* and from rotation *high lime*. The percent strontium-90 content of the soil at this depth under rotation low lime was two times higher than rotation high lime. Wiklander (5) reported that the vertical movement of strontium-90 in the soil profile was significantly higher from unlimed acid soil (pH 5.1) than from limed soil (pH 7.5).

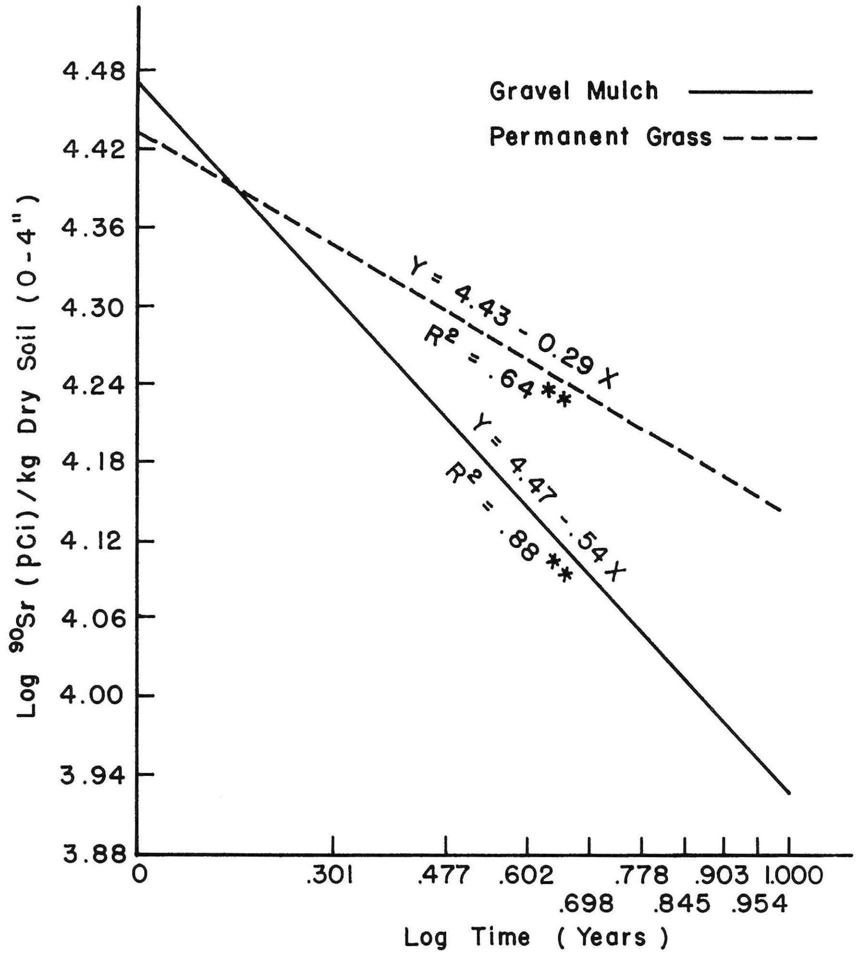

FIG. 2.—Net rate of strontium-90 removal from Canfield soil (0 to 4-inch depth) under gravel mulch and permanent grass systems.

An increase in the degree of calcium saturation favors adsorption of other cations, including strontium. Such a phenomenon can be explained on the basis of the Donnan relationship: $(Ca)_e/(Ca)_s = (Sr)_e/(Sr)_s$, where the elements in parentheses with e and s represent exchangeable and soil solution activities, respectively. When lime is added to neutralize an acid soil, $(Ca)_e/(Ca)_s$ is increased, strontium adsorption is induced, and $(Sr)_e/(Sr)_s$ is increased. In other words, the solubility of Sr in the soil is depressed. However, when excess amounts of lime are added, a gradual decrease of $(Ca)_e/(Ca)_s$ and increase in the solubility of Sr occur due to the formation of $Ca(HCO_3)_2$. Thus the adsorption of strontium is pH dependent and is least at low pH and maximum at slightly acid to slightly alkaline.

STRONTIUM-90 IN RUNOFF AND LEACHATE

The net rate of percent strontium-90 removal from microplots by runoff and leachate under different systems during a 10-year period was determined. The relationship between the log of percent strontium-90 and log time was linear for both runoff and leachate water under all cropping systems. The percent of the applied strontium-90 removed by runoff decreased with time, while its removal by leachate water increased.

The analysis of variance of percent strontium-90 in runoff water as influenced by time and management practices showed significant F values for time and cropping system. The percent strontium-90 in the runoff was considerably higher during the first year of operation than during any of the following 9 years. During the first year, strontium-90 was concentrated more on the soil surface, which in turn was subjected to more surface runoff than during the following years. The percentages of the applied strontium-90 removed by runoff under different cropping systems in descending order were: continuous corn = gravel

TABLE 4.—Total Runoff and Leachate Water (Inches) and Strontium-90 Loss (Percent of Applied) During 1962-1972 from Microplots Under Different Cropping Systems.*

Cropping System	Runoff Water, Inches†	Percent ^{90}Sr Loss	Leachate, Inches	Percent ^{90}Sr Loss
Rotation low lime	37.98	4.50	77.12	5.99
Rotation high lime	33.54	2.26	74.61	1.77
Gravel mulch	54.02	6.93	164.37	14.26
Permanent grass	19.23	1.53	89.12	2.13
Continuous corn	57.95	7.64	59.88	3.09

*Average of three replications.
†Total precipitation from June 21, 1962 to May 9, 1972 = 334.93 inches.

mulch > rotation low lime > rotation high lime = permanent grass (Table 4).

The analysis of variance of percent strontium-90 in leachate water yielded significant F values for time and cropping system. The order of percent removal of the applied strontium-90 by leachate water under different systems was: gravel mulch > rotation low lime > continuous corn > permanent grass = rotation high lime.

During a 10-year period, the percent strontium-90 loss by surface runoff was directly related to the volume of water which ran off the microplots (Table 4). On the other hand, for leachate water, the percent strontium-90 loss was not in direct proportion to the volume of water percolated through the soil profile (Table 4). This is evident when the ratio of total leachate water from gravel mulch to total leachate from permanent grass treatments (1.84) is compared with its corresponding ratio of percent loss of strontium-90 (6.70).

In addition to cropping systems, liming had a pronounced effect on the loss of strontium-90 by leaching. Such effect is noted by comparing the ratio of total leachate volume from rotation low lime to total leachate volume from rotation high lime (1.03) with its corresponding ratio of percent loss of strontium-90 (3.38). This is in agreement with the work of others (4) who have reported that acidic solutions are more effective for leaching strontium from soil surface than solutions of calcium salts. Leaching waters from the more acidic soils should release strontium-90 more readily into the drainage water than those from less acid soils (3).

In order to determine the forms of strontium-90 moving in water, runoff water from gravel mulch plots was concentrated and analyzed in a Bradfield Cell for movement of strontium-90 components in an electric field.[3] The cells were separated by a 48A pore size dialysis membrane. A low current flow was used to reduce electrolysis. The membrane was changed twice a week to minimize the effect of clogging the pores, and the solution was removed daily from the cathode and anode compartments. The volume of solution was reduced by evaporation and aliquots were taken for analysis.

The results in Table 5 indicate that equal amounts of strontium-90 moved to the cathode and to the anode. The movement of strontium to the anode indicates that it is present as a negatively charged complex. The anode material was dried and fractionated into water soluble and HCl (5%) soluble fractions. The specific activity of the HCl soluble fraction (Table 5) indicates that the strontium-90 was associated with the HCl soluble fraction.

[3]The laboratory work of this part was done by R. Shufeldt as part of his Ph.D. program at The Ohio State University.

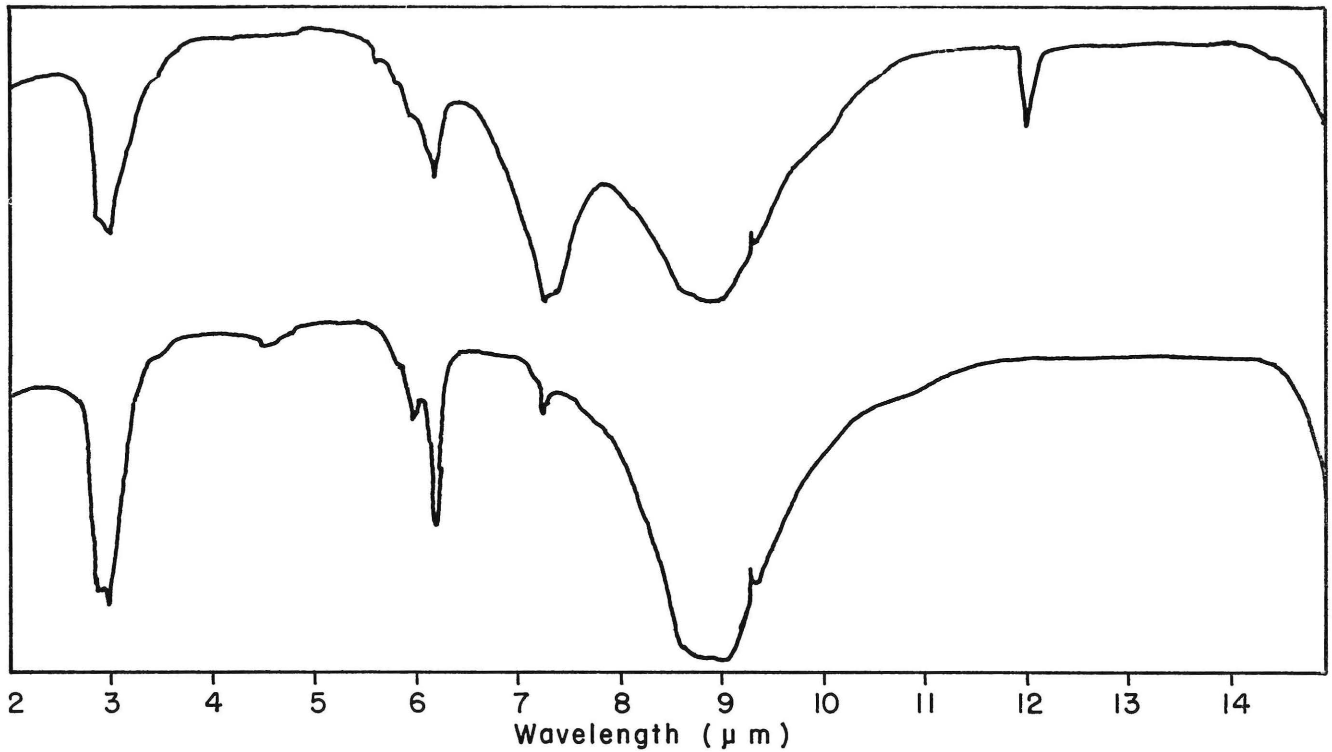

FIG. 3.—I. R. Spectra of two fractions of anode material from electrodialysis of runoff water. Total material = top spectrum and HCl soluble material = bottom spectrum.

Another factor involved in the movement of strontium in the cell could be the acidic pH of the anode compartment. The I. R. spectra of the anode material (total) and HC1 soluble fraction (Fig. 3), and the peak assignments for HC1 soluble fraction (Table 5) were determined. The spectrum of the HC1 soluble material is similar to fulvic acid spectra and to the micronutrient complexes reported by Geering and Hodgson (1), except that the hydroxyl peak is split into two peaks indicating —NH or —NH_2 groups.

PLANT UPTAKE OF STRONTIUM-90

The total yield of dry matter produced and the amount of strontium-90 removed by various crops during a 10-year period are shown in Table 6. The percent strontium-90 uptake by crops under different cropping systems, in descending order, was rotation high lime > rotation low lime > permanent grass > continuous corn.

A marked decrease in the $^{90}Sr/Ca$ ratio (PCi ^{90}Sr/meq Ca) in various crops was observed due to liming (Table 7). Since the addition of lime to an acid soil lowers the solubility of strontium, and also due to their chemical similarity and antagonistic effects, such reduction could be expected.

Among the various crops studied, alfalfa was found to be the highest accumulator and corn the least accumulator of strontium-90.

SUMMARY

The results of percent strontium-90 removed by various processes (runoff, leachate, and plant uptake) from strontium-90-treated Canfield silt loam soil under various soil and crop management practices (June 1962 to May 1972) are summarized in Table 8. The management practices consisted of one crop rotation with high and low rates of lime, permanent grass, continuous corn, and gravel mulch with no vegetation. The movement of strontium-90 in the soil profile was greatly influenced by liming and cropping system. High rates of lime retarded the downward movement of strontium-90 and reduced the plant uptake and the removal of this isotope by leachate water.

In general, crops reduced the removal of strontium-90 in runoff and leachate water. Sod crops were more effective than cultivated crops in minimizing the loss of strontium-90. The electrodialysis of runoff water indicated that equal amounts of strontium-90 moved to the cathode and to the anode. The movement of strontium to the anode was an indication of its presence as a negatively charged complex. The I.R. spectrum of the HCl soluble fraction of the negatively charged complex was similar to fulvic acid spectra and to the micronutrient complexes.

TABLE 5.—Electrodialysis of Runoff Water.

Compartment	Radioactivity (CPM/10 ml.)	Fractionation of Anode Material (3 ml. solvent/0.1 g. anode material) Fraction	Wt. (g.)	CPM/g.
Cathode	262	H_2O Soluble	0.4045	536
Anode	278	HCl Soluble	0.1453	1796

I. R. peaks of HCl soluble fraction

Peaks (microns)	Assignment
2.85-3.0 Double	—NH or —NH_2
5.95	C = O
6.20	C = O
8.5-9.0 Broad	
15.0	Chloride

TABLE 6.—Yield of Dry Matter Produced and Strontium-90 (Percent of Applied) Removed by Crops During 1962-1972.

Cropping System	Total Yield, Kg.	Percent ^{90}Sr Removed
Rotation low lime	48.94	11.30
Rotation high lime	63.38	14.11
Permanent grass	40.71	10.08
Continuous corn	95.83	8.02

TABLE 7.—^{90}Sr:Ca Ratio in Crops as Influenced by Liming.

Treatment	Corn		Wheat			Alfalfa
	Fodder	Shelled Corn	Straw	Chaff	Grain	
	pCi ^{90}Sr/meq Ca					
Rotation low lime	128	90	213	161	133	137
Rotation high lime	88	64	156	126	109	88

TABLE 8.—Percent of the Applied Strontium-90 Removed by Runoff (Water + Sediment), Leachate Water, and Crops from Treated Microplots (June 1962 to May 1972).

Cropping System	Runoff	Leachate	Crops	Total
	Percent ^{90}Sr			
Gravel mulch	6.93 a*	14.26 a		21.18
Rotation low lime	4.50 b	5.99 b	11.30 b	21.79
Continuous corn	7.64 a	3.09 c	8.02 d	18.75
Rotation high lime	2.26 c	1.77 d	14.11 a	18.14
Permanent grass	1.53 c	2.13 cd	10.08 c	13.74

*Means followed by the same letter are not significantly different at 5% level.

The quantities of strontium taken up by the various crops differed and such differences among species roughly paralleled their absorption of calcium. Strontium-90 removal by crop uptake from all cropping systems with the exception of the continuous corn system was significantly higher than from their corresponding runoff and leachate water combined.

Ten-year data from the field microplots indicate that the behavior of strontium-90 in Canfield silt loam under natural environmental conditions is markedly affected by soil and crop management practices.

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Green Springs Crops Research Unit, Green Springs, Sandusky County: 26 acres

Jackson Branch, Jackson, Jackson County: 344 acres

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Muck Crops Branch, Willard, Huron County: 15 acres

North Central Branch, Vickery, Erie County: 335 acres

Northwestern Branch, Hoytville, Wood County: 247 acres

Pomerene Forest Laboratory, Keene Township, Coshocton County: 227 acres

Southeastern Branch, Carpenter, Meigs County: 330 acres

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Western Branch, South Charleston, Clark County: 428 acres